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Theoretical and experimental investigations of optical diffraction on patterned surface

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Abstract

Optical diffraction is widely used to probe micrometre sized patterns given the wavelength of optical light. However, the theoretical background underlying the observed diffraction patterns is rich and can be not trivial. The investigation of this background associated with an experimental work on patterned surface allow to perform computational simulation of diffraction phenomena, providing a deep understanding of the latter. Two types of samples were studied during this project course, leading to both analytical and computational studies of optical diffraction.

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Chapter 1

Introduction

Materials sciences and solid state physics are the main domains of physics where the dimensions and the typical size of the studied systems are of major importance. In the field of nanosciences, condensed matter systems offer properties that are very dependent on their size, when the surface of the system dominates over its volume. At the nanoscale, matter can be probed with light of wavelength of same order of magnitude, that is mostly X-rays. However, building nanostructures and patterned surfaces at the nanoscale can be involved and technically demanding. The study of matter at the atomic scale is subjected to surface cleaning since it is also the scale of chemistry and therefore requires most often vacuum or even ultra-high-vacuum conditions, as well as the use of X-Rays.

The understanding and characterization of patterned surface using diffraction methods for topography purposes can be performed at a larger length scale, whenever the classical limits are still valid, that is to say use models of light scattering in classical electrodynamics. Such approach can be performed on the study of micrometre scaled patterned surface with the help of visible light. Highly collimated light sources such as lasers in the visible range have wavelength compatible with such materials.

In this project course, optical diffraction is investigated experimentally on two types of patterned samples. An analytical and computer-based simulations approaches are also adopted and compared with the experiment, in order to try to understand the physics of optical diffraction in terms of light scattering thanks to complementary methods.

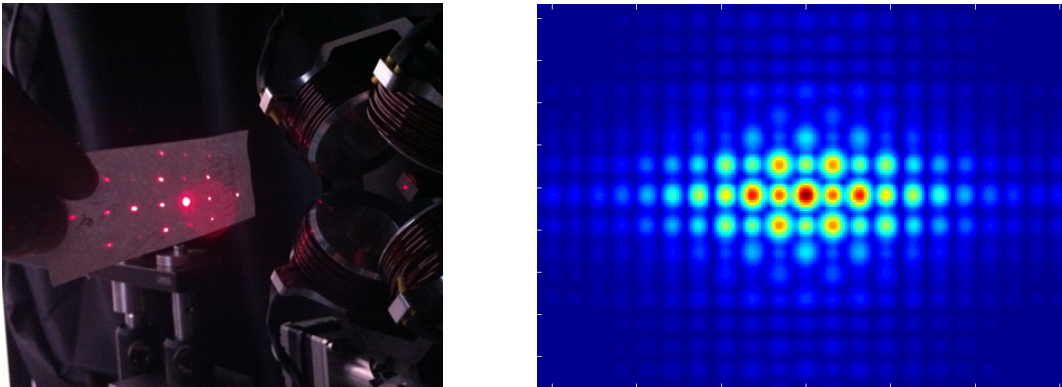


Figure 1.1: *Photograph of an optical diffraction pattern obtained experimentally, along with a picture of the corresponding computer simulated diffraction pattern.*

Chapter 2

Theoretical basis and experimental setup

The aim of this chapter is to bring up to the reader a global overview of both theoretical and experimental tasks that I have been doing during this project course. The first part describes some basic elements of the optical scattering theory, from crystal structure to patterned surface. The second part shows the experimental equipment and set-up used to measure and gather optical diffraction patterns from the scattering of laser light onto a patterned surface, and how the diffraction patterns have been explored within this project course.

2.1 Elements of optical scattering theory on patterned surface

2.1.1 Optical scattering theory and kinematic approximation

The origin of the word diffraction is attributed to Francesco Maria Grimaldi [1], who used the Latin word *diffringere* which means to break into pieces, to describe the behaviour of light going through different media. Within the same century, James Gregory [2] observed the fashion that light passed through a bird feather, therefore discovering the first diffraction grating. In 1801, Thomas Young performed his nowadays famous two slits experiment, describing how light interferes to produce specific patterns. This experiment gave, with many others, great support to the wave theory of light that had been advanced by Christiaan Huygens [3] who stated, together with Augustin Fresnel [4], that each point of an advancing wave front can be regarded as the source of a secondary train of waves. The advancing wave as a whole may then be described as the sum of all the secondary waves arising from all points already traversed by the wave.

Indeed light as an electromagnetic wave can be fully described by its electric field in a specific medium thanks to Maxwell's equations. Therefore, the study of optical interaction of light through matter can be fully done with the description of the electric field. Two fundamental aspects are covered: the change of the incident electromagnetic wave in interaction with matter and the calculation of interferences of the scattered waves.

Hence two fundamental assumptions about the scattering process lead to what is called the kinematic approximation. Firstly the scattering is considered to be elastic, meaning that the wavelength of the scattered wave is equal to the one of the incident one, or in other word the scattered photon maintains the same energy as the incoming one. Secondly, the *strength*

of the scattering, the scattering cross-section α is assumed to be small. The probability for light to scatter twice is of order α^2 which is very much smaller than for single scattering. The contributions of multiple scattering are neglected under those circumstances and justify the truncation at the first term. This is the so called *first Born* or *single scattering approximation*.

In the following, one describes the scattering amplitude from a single electron, then from atoms and groups of atoms and finally from patterned surface wherein surface diffraction occurs.

2.1.2 Scattering from electrons to charge distribution

A single electron can be viewed as a classical point charge. The interaction with an incoming electromagnetic wave of wave vector $\mathbf{k}$ forces this point charge to oscillate with the same frequency $\omega = kc$ as the incoming wave where c is the speed of light in vacuum. The plane in which the electron oscillates is given by the direction of polarization of the electric field. According to classical electrodynamic theory, the accelerated point-charge radiates. The resulting radiation is emitted from the oscillating electron and has the same frequency ω as the exciting primary wave. This process is called *scattering* or, more precisely, *Thomson scattering*.

The incident wave can be considered as a plane wave in the far field approximation. With an amplitude A_0 and propagating along the wave vector $\mathbf{k}$, it can be written $A_0 e^{-i\mathbf{k}\cdot\mathbf{r}}$. The scattered wave originating from the interaction between the incident wave and the free electron located at position $\mathbf{r}$ is written¹

$$A_e e^{-i\mathbf{k}'\cdot\mathbf{r}} = A_0 e^{-i\mathbf{k}\cdot\mathbf{r}} \frac{1}{4\pi\epsilon_0} \frac{e^2}{mc^2} \frac{1}{R} p. \quad (2.1)$$

The scattered wave travels in the direction of $\mathbf{k}'$ with the amplitude A_e and is observed at a distance R from the interaction point.

As described above, the scattered wave corresponds to the radiation that originates from the accelerated oscillation of the probed free electron. As a matter of fact, the scattered amplitude in the plane defined by the vectors $\mathbf{k}$ and $\mathbf{k}'$ (i.e., the scattering plane) varies with the direction of this oscillation, which is determined by the direction of polarization of the light in use. The projection of the acceleration is proportional to $\cos\theta$. Since we have used *s*-polarized light, the scattering is perpendicular to the polarization plane of the electric field. The projection of the acceleration of the oscillation of the electron is null on the scattered plane and the scattered wave is therefore *s*-polarized. There is no modulation of the amplitude of the scattered wave in the scattering plane with respect to the polarization since it is perpendicular and $p = 1$.

Substituting $\frac{1}{4\pi\epsilon_0} \frac{e^2}{mc^2} = r_e$ and $A_0 \frac{r_e}{R} p = \mathcal{K}$ and $\mathbf{q} = \mathbf{k}' - \mathbf{k}$, one obtains

$$A_e = \mathcal{K} e^{i\mathbf{q}\cdot\mathbf{r}}. \quad (2.2)$$

This scattering contribution from a single free electron can be extended to a continuous charge distribution by integrating over space a charge density $\rho(\mathbf{r}')$, that can represent the electronic distribution for a single atom, as follows

¹The following derivation is greatly inspired from the work of Christian M. Schlepütz [5].

$$\begin{aligned}
A_\rho &= \mathcal{K} \int e^{i\mathbf{q} \cdot (\mathbf{r} + \mathbf{r}')} \rho(\mathbf{r}') d^3\mathbf{r}' \\
&= \mathcal{K} e^{i\mathbf{q} \cdot \mathbf{r}} \int e^{i\mathbf{q} \cdot \mathbf{r}'} \rho(\mathbf{r}') d^3\mathbf{r}'.
\end{aligned} \tag{2.3}$$

2.1.3 Scattering from electrons to an atom

One now considers a single atom. It is characterized by a relatively (with respect to the electrons) small and heavy nucleus of protons around which electrons are described by a charge distribution $\rho_a(\mathbf{r}')$. From the result of subsection 2.1.2 the scattering contribution of the atom is given by

$$\begin{aligned}
A_a &= \mathcal{K} e^{i\mathbf{q} \cdot \mathbf{r}} \int_{\text{atom}} e^{i\mathbf{q} \cdot \mathbf{r}'} \rho(\mathbf{r}') d^3\mathbf{r}' \\
&= \mathcal{K} e^{i\mathbf{q} \cdot \mathbf{r}} f_a(\mathbf{q}),
\end{aligned} \tag{2.4}$$

where

$$f_a(\mathbf{q}) = \int_{\text{atom}} e^{i\mathbf{q} \cdot \mathbf{r}'} \rho(\mathbf{r}') d^3\mathbf{r}' \tag{2.5}$$

is called the *atomic form factor* and is equal to the Fourier Transform of the atomic electron density.

2.1.4 Scattering from isolated atoms to unit cell

Atoms can rearrange in a regular manner to form crystalline solids. The work of the crystallographer is mainly to probe the crystalline structure with the scattering of radiations having the appropriate wavelength consistent with the order of magnitude of the crystal structure.

First, let us consider a single unit cell at position $\mathbf{r}_{uc}$ in space, containing N atoms identified by an index j . Each atom has an atomic form factor $f_a(\mathbf{q})$ and is located at position $\mathbf{r}_j$ in the unit cell. The scattering of the whole unit cell is the coherent addition of the scattering contribution of each individual atoms it contains

$$\begin{aligned}
A_{uc} &= \mathcal{K} \sum_{j=1}^N f_j(\mathbf{q}) e^{i\mathbf{q} \cdot (\mathbf{r} + \mathbf{r}_{uc} + \mathbf{r}_j)} \\
&= \mathcal{K} e^{i\mathbf{q} \cdot \mathbf{r}} e^{i\mathbf{q} \cdot \mathbf{r}_{uc}} \sum_{j=1}^N f_j(\mathbf{q}) e^{i\mathbf{q} \cdot \mathbf{r}_j} \\
&= \mathcal{K} e^{i\mathbf{q} \cdot \mathbf{r}} e^{i\mathbf{q} \cdot \mathbf{r}_{uc}} F(\mathbf{q}).
\end{aligned} \tag{2.6}$$

As it has been done for the atomic form factor, one can combine all the phase factors associated with the atoms in the unit cell into a single $\mathbf{q}$ -dependent term $F(\mathbf{q})$, called the *structure factor*. This term contains the information of the position of the atoms in the unit cell as well as their type laying in the corresponding atomic form factor. It corresponds² to the

²In equation 2.7, n is the number of different types of atoms in the unit cell

sum of the individual atomic form factors $f_j(\mathbf{q})$ weighted with the corresponding phase shift given the various positions of atoms inside the whole unit cell

$$F(\mathbf{q}) = \sum_{j=1}^n f_j(\mathbf{q}) e^{i\mathbf{q} \cdot \mathbf{r}_j}. \quad (2.7)$$

One can mention the expression of the scattering amplitude for a three dimensional crystalline structure composed of N_1, N_2, N_3 unit cells in the respective spatial direction a_1, a_2, a_3 . The amplitude A_c of the wave scattered by the entire crystal is then given by the amplitude A_{uc} from a single unit cell multiplied by the sum over the phase terms associated with the unit cell position $\mathbf{r}_{uc} = j_1 \mathbf{a}_1 + j_2 \mathbf{a}_2 + j_3 \mathbf{a}_3$, usually referred to as the *lattice sum* (where the j_i coefficients identify the unit cell's position).

$$A_c = \mathcal{K} e^{i\mathbf{q} \cdot \mathbf{r}} F(\mathbf{q}) \sum_{j_1=0}^{N_1-1} e^{i\mathbf{q} \cdot (j_1 \mathbf{a}_1)} \sum_{j_2=0}^{N_2-1} e^{i\mathbf{q} \cdot (j_2 \mathbf{a}_2)} \sum_{j_3=0}^{N_3-1} e^{i\mathbf{q} \cdot (j_3 \mathbf{a}_3)}. \quad (2.8)$$

One can see geometric series in each sum and therefore the amplitude can be rewritten more explicitly. One describes the derivation in appendix A.1. The scattering amplitude of the ideal 3-dimensional crystal is then written

$$A_c = \mathcal{K} e^{i\mathbf{q} \cdot \mathbf{r}} F(\mathbf{q}) \frac{\sin(\frac{1}{2} N_1 q_1 a_1)}{\sin(\frac{1}{2} q_1 a_1)} e^{iq_1 a_1 \frac{N_1-1}{2}} \frac{\sin(\frac{1}{2} N_2 q_2 a_2)}{\sin(\frac{1}{2} q_2 a_2)} e^{iq_2 a_2 \frac{N_2-1}{2}} \frac{\sin(\frac{1}{2} N_3 q_3 a_3)}{\sin(\frac{1}{2} q_3 a_3)} e^{iq_3 a_3 \frac{N_3-1}{2}}, \quad (2.9)$$

and one uses it in the following section when expressing the scattering amplitude for surface diffraction. As a matter of fact, this expression of the scattering amplitude of the whole crystal can be used as a starting point for the study of surface diffraction, where the crystal may be truncated in one direction (the out-of-plane direction a_3 by convention).

2.1.5 Surface reflective diffraction

The aperiodicity of a finite crystal close to its surface leads to consider the unit cells from $-\infty$ to 0 in the a_3 -direction. Therefore, the scattering contribution A_s of such a crystal *slab* differs slightly from the scattering amplitude obtained for an finite 3-dimensional crystal in equation 2.8, in the sum over j_3 in the a_3 -direction. Before the transformations in terms of geometric sums, the scattering amplitude of the surface reads

$$A_s = \mathcal{K} e^{i\mathbf{q} \cdot \mathbf{r}} F(\mathbf{q}) \sum_{j_1=0}^{N_1-1} e^{i\mathbf{q} \cdot (j_1 \mathbf{a}_1)} \sum_{j_2=0}^{N_2-1} e^{i\mathbf{q} \cdot (j_2 \mathbf{a}_2)} \sum_{j_3=-\infty}^0 e^{i\mathbf{q} \cdot (j_3 \mathbf{a}_3)}. \quad (2.10)$$

The geometric sums in equations 2.8 and 2.10 can be transformed (see appendix A.1 on page 20) in order to express a nice expression of the diffracted intensity. In fact, the physical quantity collected when studying diffraction patterns is the *intensity* of the scattered light. It is defined as the modulus squared of the complex electric field of the outgoing wave, expressed in terms of scattering amplitude in this present work. Hence, one has therefore the intensity as a function of the scattering vector $\mathbf{q}$

$$I_s(\mathbf{q}) = I_0 \frac{r_e^2}{R^2} |F(\mathbf{q})|^2 \frac{\sin(\frac{1}{2} N_1 q_1 a_1)}{\sin(\frac{1}{2} q_1 a_1)} \frac{\sin(\frac{1}{2} N_2 q_2 a_2)}{\sin(\frac{1}{2} q_2 a_2)} \frac{1}{4 \sin(\frac{1}{2} q_3 a_3)}. \quad (2.11)$$

2.1.6 Focus on the structure Factor $F(\mathbf{q})$

Within this work, a derivation of the intensity has been understood and given as an analytical basis for the experimental work presented in the next chapter. In order to present a whole consistent work that merges experimental, analytical and computational works, the choice of focusing on the structure factor $F(\mathbf{q})$ has been made for the sake of simplicity since it contains all the information that characterizes the studied samples.

Also, we are dealing with optical diffraction and therefore we use visible light to probe our samples. Such light has a wavelength of order of magnitude 10^{-1} micrometre, compatible for diffraction with structures of the same order of magnitude. Our samples are patterned surface of alloys, arranged on structure of the size of micrometre. Therefore, one can use the same equations as for x-ray or electronic diffraction on crystals but applied on micrometre sized patterned surface, for our purposes of optical diffraction. The atomic form factor can describe not only matter at the atomic scale but also, at a much larger scale, matter in terms of solid structure of thousands of atoms.

2.2 Setup for optical reflective diffraction measurements

2.2.1 Experimental setup

The experimental work has consisted in collecting optical diffraction pattern reflected by the diffracting samples in study. The experimental setup depicted in figure 2.1 is made of a laser of wavelength 660 nm (660-100 diode laser, Cube) fixed on an optical bench containing a polariser and an iris. The laser beam propagates in s-mode after the polariser and its diameter decreases as it passes the iris. It is then reflected and diffracted by the sample which is attached to a doubly rotatable stage. The diffracted beams are collected by a detector attached to the rotatable stage and positioned so that it forms with the laser beam a horizontal plane (which is actually the scattering plane).

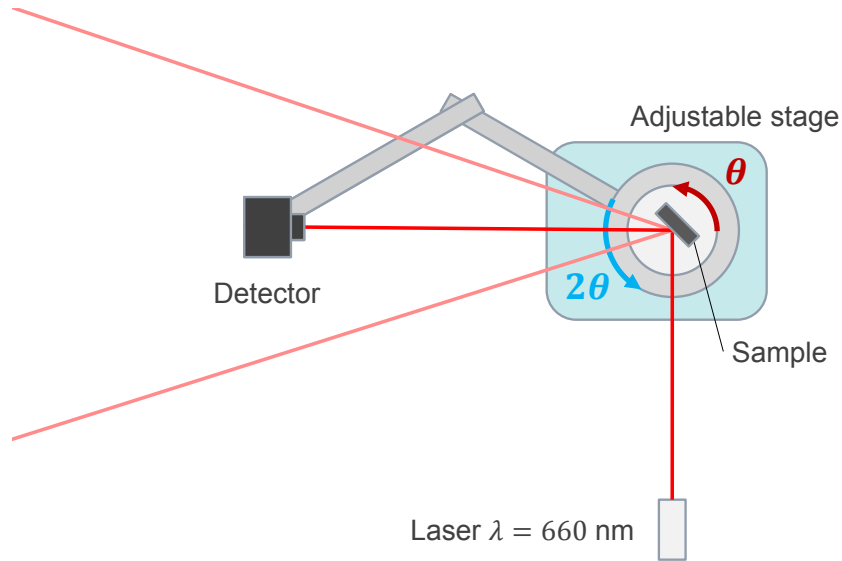


Figure 2.1: Schematic view of the experimental set-up seen from above, showing the incident laser beam diffracted by the sample. Diffracted light is collected by the detector positioned perpendicularly to the incident beam.

The measurements were performed with a fixed detector positioned perpendicularly to the direction of the incident beam ($2\theta = 90^\circ$) and by rotating the sample along the angle θ . The rotatable stages are moved by a command-line programme which directs the rotations of the two circles goniometer. A scan starts by defining the desired angular range for the sample to be rotated in, and by setting the number of steps, i.e., the angular resolution, for the detector to collect the intensity. Therefore, a measurement ends up with a diffraction pattern in terms of light intensity as a function of the rocking angle θ .

2.2.2 From measurements to computation

In this work, the measured surface diffraction patterns were done for known samples. The geometry of the studied surfaces is known prior to the measures. The collected diffraction patterns were not used to characterize the samples but in a more general way to study and understand optical surface diffraction, from experimental, analytical and computational points of view. As a matter of fact, one started with a known simple specific patterned surface, made measurements as well as analytical calculations (for predictions and verifications) and finally computer simulation, by building the sample and its diffraction virtually. These three approaches, already mentioned earlier, represent the essence of this short study of optical surface diffraction.

Chapter 3

Study of a patterned surface made of stripes of alternate width

In order to cover experimental, analytical and computational tasks, one has decided to work with a simple patterned surface made of stripes of alternative width, simple enough for the diffraction pattern to be measured, calculated and simulated correctly.

3.1 Measurements and data treatment

The sample is an inverse Permalloy $\text{Ni}_{20}\text{Fe}_{80}$ with alternating 200 nm and 400 nm stripes such that a 2 dimensional unit cell can be defined along the x direction as shown in figure 3.1. The corresponding lattice parameter, i.e., the spatial frequency, is called a .

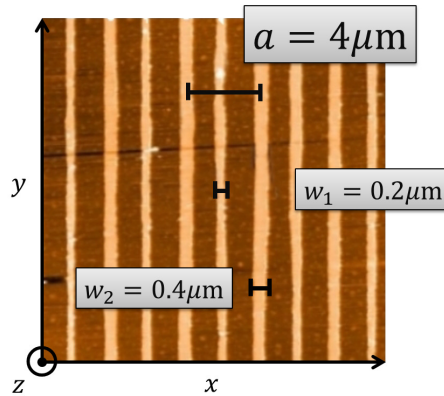


Figure 3.1: Atomic Force Microscopy picture of the sample made of stripes of alternate width, in the out-of-plane direction.

3.1.1 Experimental measurements

It is placed on its rotatable holder, ready for a rocking scan (θ scan). The detector is at a fixed position during the scanning process, at approximately one metre away from the sample, forming an angle of 90° with the incident beam. Two measurements were performed, the first being one fifth as resolved as the second, and are presented in figure 3.2, respectively 3.2a and 3.2b.

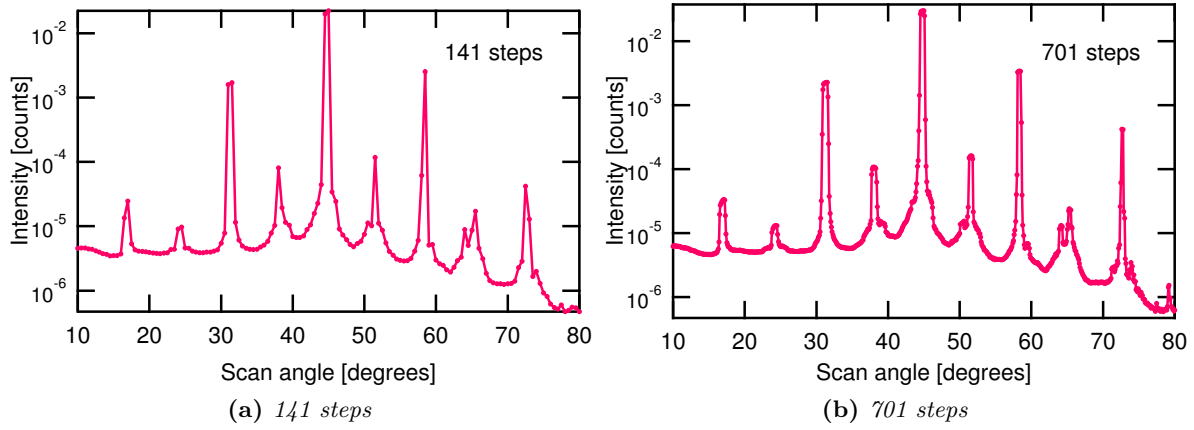


Figure 3.2: Diffraction patterns (intensity versus scattering angle θ) for two 2θ scans. The two scans have been made by rotating the sample from the angle $\theta = 10^\circ$ to $\theta = 80^\circ$, in respectively 141 steps (i.e., with 0.5° of accuracy) and 701 steps (i.e., with 0.1° of accuracy).

In spite of the dark blanket that protected the experimental setup from external light, some light background has been gathered by the detector while scanning. Indeed, the ideal shape of the curve for intensity as a function of θ should lay down on zero with sharp and high value at diffraction peaks. This background originates from the remaining ambient light in the room and from imperfections of the structure which rise to diffuse scattering.

Since, in the following, one will focus on analytical and computational results and compare them with the experiment, one can wisely wonder how the relative vagueness of the measurements can be reduced to match better with the two more ideal-like approaches that are the analytical and computational ones.

3.1.2 Data treatment: background reduction

A specific treatment of the collected data has thus been elaborated in this work, to reduce this undesired gathered background light. Considering the intensity of this background as the region under the intensity peaks, a MATLAB function has been written whose purpose is to subtract the background from the actual diffracted light intensity, for each peak of any diffraction pattern. Taking as an input the diffraction pattern shown in figure 3.2, the routine individualizes the peaks and subtracts linearly the background step by step, to flatten the peaks down to zero. Figure 3.3 depicts the nine measured peaks (i.e., containing the light signal of the diffracted beams plus the background) in blue, and the corrected ones are shown in black (without the considered background).

In order to compare the analytical and computational approaches to the measurements, this treatment of the data is useful when considering the *integrated intensity*, which will be treated in the following. The first two mentioned approaches do not take into account any background, which shows that this data treatment is rather purposeful. However, powerful automatized routines can easily be found and would replace and surpass this home-made function. This was designed more to handle MATLAB and its features than to actually develop a reusable sturdy routine.

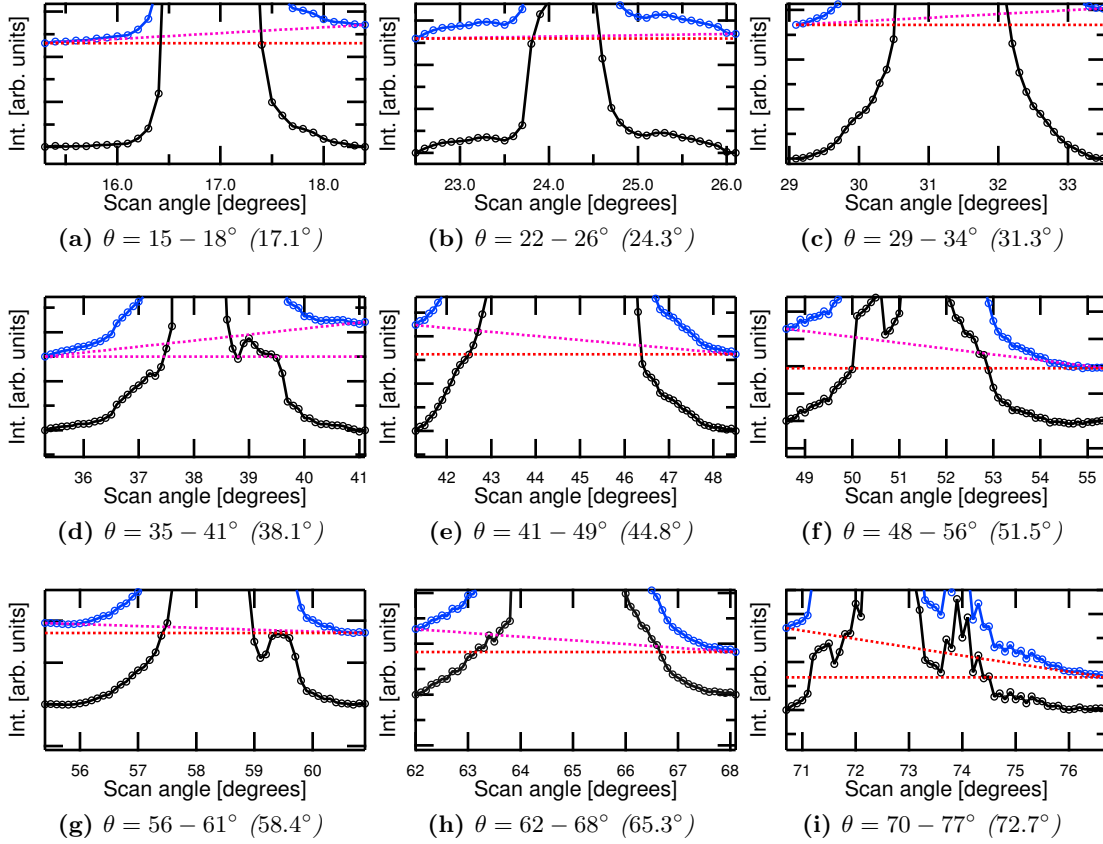


Figure 3.3: Diffraction peaks treated from the pattern of figure 3.2b where a linear background correction has been applied. A MATLAB routine takes the minima between every peaks and applies a linear subtraction individually for each one of them. The blue lines correspond to the untreated data, whereas the black ones represent the background subtracted peaks. For each scan angle value (x axis), the experimental intensity (y axis) is lowered by an amount of intensity which is equal to the difference between the intensity at the pink dotted line with the intensity at the red dotted line. This represent a linear background reduction.

3.2 Analytical investigation

After dealing with the experimental approach, one presents the analytical investigation of optical diffraction for the sample of alternating width stripes. It corresponds to an application of the theoretical part covered in the first chapter for the actual grating structure in study.

3.2.1 Building the sample in the real space

The sample made of stripes of alternate width can be viewed as a particular grating structure. It can be described as infinitely periodic in the x direction, infinitely symmetric in the y direction, and finite in the z direction. Therefore, the directions of interest are the *in-plane* x direction and the *out-of-plane* z direction. The actual topography of the surface in these directions can be mathematically viewed as a sequence of *rectangular functions* of alternate width.

The surface unit of the 2-dimensional lattice in study can be modelled as the sum $\rho(\mathbf{r})$ of two distributions $\rho_1(\mathbf{r})$ and $\rho_2(\mathbf{r})$, representing the *thin* and *thick* stripes respectively, in the

(x, z) plane. In a unit cell¹ of lattice parameter a , the width of the thin and thick stripes is called w_1 and w_2 respectively, and the their distribution in space is expressed in the following equations

$$\rho_1(\mathbf{r}) = \begin{cases} 1 & \text{if } \begin{cases} |x| \in [0 ; \frac{w_1}{2}] \\ 0 \leq z \leq h \end{cases} \\ 0 & \text{otherwise} \end{cases} \quad (3.1a)$$

$$\rho_2(\mathbf{r}) = \begin{cases} 1 & \text{if } \begin{cases} |x| \in [\frac{a-w_2}{2} ; \frac{a}{2}] \\ 0 \leq z \leq h \end{cases} \\ 0 & \text{otherwise} \end{cases} \quad (3.1b)$$

$$\rho(\mathbf{r}) = \rho_1(\mathbf{r}) + \rho_2(\mathbf{r}). \quad (3.1c)$$

The unit slab can be represented as shown in figure 3.4, which is a cut on the (x, z) plane of the actual sample which can be observed on the AFM picture in figure 3.1 in section 3.1, in which the contrast depicts the two types of stripes of non-zero height and of alternative width, arranged periodically.

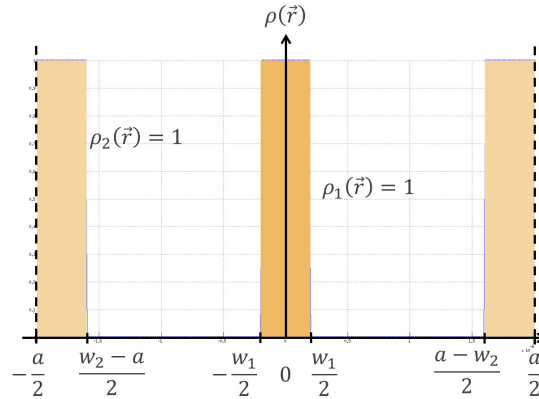


Figure 3.4: Plot of a side view (plane (x, z)) of the surface unit slab depicted as the sum of two distributions $\rho_1(\mathbf{r})$ and $\rho_2(\mathbf{r})$, as defined in expressions 3.1

According to section 2.1.6 which emphasizes on the importance of the structure factor $F(\mathbf{q})$ as defined in equation 2.7, the mathematical derivation of diffraction patterns in terms of intensity with respect to the angle θ (or equivalently as a function of the scattering vector $\mathbf{q}$) requires the atomic form factors² and therefore solely the knowledge of the distribution $\rho(\mathbf{r})$, which are known as defined in equation 3.1c and the associated ones (3.1a and 3.1b).

3.2.2 Calculation of the structure factor and diffracted intensity

The structure factor $F(\mathbf{q})$ is defined as the sum of the atomic form factors weighted by a phase factor that takes into account the relative position of the distributions in the unit slab. As shown in figure 3.4, the two distributions $\rho_s(\mathbf{r})$ ($s = 1, 2$) are centred in the origin of the x axis. Therefore, their position vector $\mathbf{r}_s$ are both null i.e. $\mathbf{r}_1 = \mathbf{r}_2 = \mathbf{0}$. From this follows

¹As discussed in section 2.1.5, one shall use the term *unit slab* since we are now dealing with a surface

²See equation 2.5.

that the phase factors $e^{i\mathbf{q}\cdot\mathbf{r}_s}$ are unitary and the structure factor expressed in equation 2.7 becomes

$$F(\mathbf{q}) = \sum_{s=1}^2 f_s(\mathbf{q}). \quad (3.2)$$

The unit slab under investigation is mathematically represented by two spatial distributions $\rho_s(\mathbf{r})$, one for each stripes. Thus the two form factors $f_s(\mathbf{q})$ are defined as the spatial Fourier Transform over x and z coordinates of the corresponding distribution, as expressed below

$$\begin{aligned} f_1(\mathbf{q}) &= \int \rho_1(\mathbf{r}') e^{i\mathbf{q}\cdot\mathbf{r}'} d^3\mathbf{r}' \\ f_2(\mathbf{q}) &= \int \rho_2(\mathbf{r}') e^{i\mathbf{q}\cdot\mathbf{r}'} d^3\mathbf{r}'. \end{aligned} \quad (3.3)$$

The structure factor hence writes

$$F(\mathbf{q}) = \int \rho_1(\mathbf{r}') e^{i\mathbf{q}\cdot\mathbf{r}'} d^3\mathbf{r}' + \int \rho_2(\mathbf{r}') e^{i\mathbf{q}\cdot\mathbf{r}'} d^3\mathbf{r}'. \quad (3.4)$$

Calculating the structure factor $F(\mathbf{q})$ is done by deriving the expression of the two form factors. It consists in calculating the 2 dimensional Fourier Transforms of the distributions $\rho_1(\mathbf{r})$ and $\rho_2(\mathbf{r})$. The analytical derivation is presented in appendix A.2 on page 21.

The structure factor leads to the diffracted intensity as seen in equation 2.11 since the latter is proportional to the square modulus of the former. This provides an analytical expression for the intensity, that we compare in the following to computational approach based on simulation.

3.3 Simulation as a theoretical experiment

The last approach to be presented is the simulation of optical diffraction. The sample presented in the previous section is computerized in the sense that the geometry of the striped sample is reproduced in MATLAB. Given the symmetry of the sample, which is infinitely symmetric along the y direction and where the unit slab runs periodically in the x direction, we use a two-dimensional model where the stripes are represented by periodic step functions, as shown in figure 3.4 in section 3.2 on page 11 dealing with the analytical approach. The two topological quantities in play are the width of the striped surface and the height of the stripes. They are represented by the x and z axis respectively. These two directions lie in a 2 dimensional matrix which can be seen as a map of the sample. Therefore, the matrix elements represent the atomic distribution $\rho(\mathbf{r})$ as written in equation 3.1c. The computed matrix is thus filled with zeros and ones, whether the distribution is one or zero if it falls on a stripe or not.

The diffracted intensity defined in equation 2.11 is proportional to the square modulus of the structure factor. According to the symmetry of the problem, the structure factor is nothing but the sum of the form factor $f_s(\mathbf{q})$ of the two types of stripes which are the Fourier transform of the distributions $\rho_s(\mathbf{r})$. That is why the simulated diffraction pattern can reasonably be computed taking the 2 dimensional Fast Fourier Transform of the distribution matrix.

The resulting 2-dimensional matrix contains the diffraction pattern, which corresponds to the intensity with respect to the reciprocal space coordinates. Using Bragg law, the reciprocal

space can be spanned as a function of the scanning angle θ , such that the scattering vectors' components q_x and q_z reads

$$\begin{aligned} q_x &= \frac{2\pi}{\lambda} (\cos(2\theta - \omega) - \cos \omega) \\ q_z &= \frac{2\pi}{\lambda} (\sin \omega + \sin(2\theta - \omega)). \end{aligned} \quad (3.5)$$

From the above expressions, reduced coordinates can be defined as follows

$$\begin{aligned} q_x^R &= q_x \cdot \frac{a}{2\pi} \\ q_z^R &= q_z \cdot \frac{a}{2\pi}. \end{aligned} \quad (3.6)$$

3.3.1 Generation of the diffraction pattern in the reciprocal space

The diffraction pattern that is the intensity as a function of the scanning angle θ is obtained mainly by the computation of the FFT of the distribution functions in the structure factor and the square modulus of the latter. Figure 3.5 depicts the computed structure factor with given geometric parameters (a , ω_1 , ω_2). It presents the comparison between the structure factor obtained analytically and the one obtained based on the simulated model (see distributions 3.1). The analytical calculation of the diffracted intensity (Eq. 2.11) differs from what is processed during the simulation, only in the computation of the structure factor, which is either done by hand or through FFT of the computed distributions functions. However, the starting model is the same in both cases. The two methods to calculate the diffraction pattern yield comparable results as can be seen in the figure 3.5. This validates the two different implementations.

3.3.2 Refinement of the virtual sample: matching with experiment

The intensity of the simulated diffraction pattern can reach better matching with the experimentally obtained patterns when tuning the geometric parameters ω_1 , ω_2 and the lattice parameter a of the unit slab. As we presented in the previous section, the analytical and simulation approaches are based on the same model and show good agreement together. Therefore even though we don't have access to the actual calculation performed when using the *FFT* of the distribution functions, we still have a good hint of the important parameter in play by looking at the explicit expression of the structure factor shown in appendix A.2. The geometric parameters come into play both in amplitude and in frequency since they appear as pre factor as well as argument of the cardinal sine functions. If the model is a sufficient one to mimic the actual optically investigated system, by varying the input parameters such as these geometric parameters in such a way that convergence can be found between the simulated and the experimental diffraction patterns, then we can refine the knowledge of these parameters. In other words, the simulation experiment can be used as a measurement of the sample structure, as far as we can arbitrary consider that the input model is convenient.

Following this procedure, a good matching in the relative intensity of the peaks could be found for $\omega_1 = 0.56 \mu\text{m}$, $\omega_2 = 2.00 \times \omega_1$, and $a = 4.102 \mu\text{m}$. Figure 3.6 displays the superimposition of the experimental diffraction pattern with the analytical one, but computed with the above mentioned geometric parameters' value.

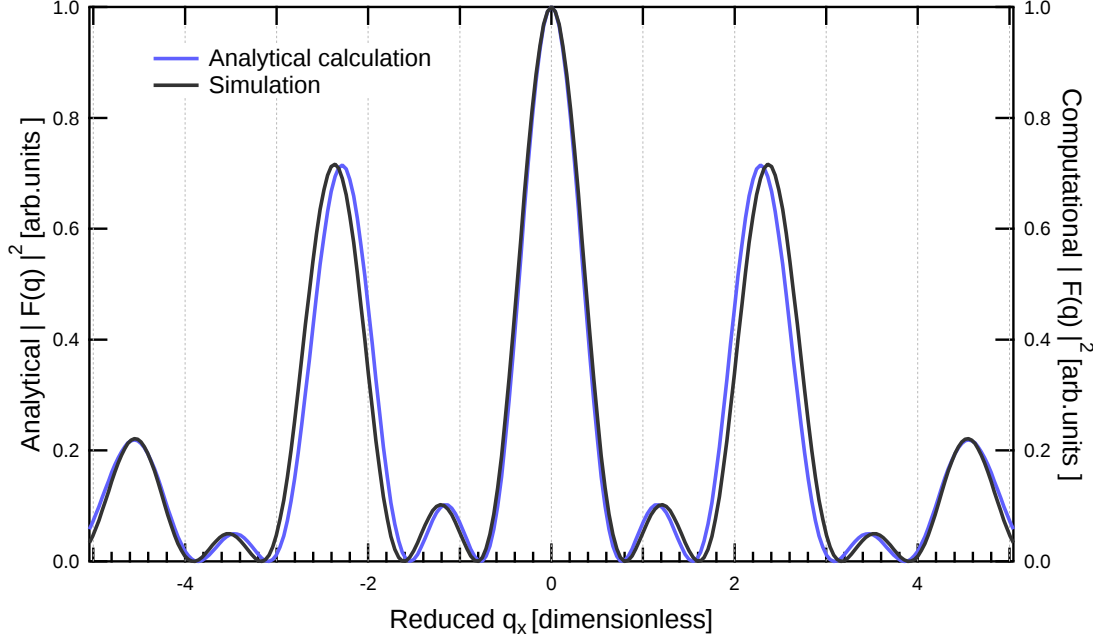


Figure 3.5: Square modulus of the structure factor within the analytical calculation (computation of $|F(\mathbf{q})|^2$ as obtained in appendix A.2, blue curve) and simulation (computation of FFT of the modelled distribution of matter, black curve) showing good matching of these two analogous approaches. The x -axis is spanned by the reduced scattering vector component q_x defined through equation 3.6 and which is the spanning variable of the reciprocal space where lies the structure factor and its Fourier transform nature.

3.3.3 Integrated intensity

In order to quantitatively compare the different approaches, one needs a physical quantity that is independent from the way it is obtained, that is only dependent on the system under study. From a rocking scan to another, according to the starting geometry of the measurement, i.e. here the scan were obtained with a fixed detector at a normal position with respect to the incident beam and by tilting the sample, the diffraction pattern can be different from one experiment to another, being a function of diffracted intensity versus the scanning angle. Moreover, the data treatment applied on the experimental diffraction pattern which gives as outputs the individual diffraction peaks can be easily followed by a calculation of the integrated intensity. The integrated intensities of the experiment and of the calculations are compared in figure 3.7 and again, good agreement is found.

3.4 Two dimensionally patterned surface: outlooks

Besides the striped sample, we have been provided a patterned sample made of spheres and ellipsoids islands, represented in figure 3.8a by a picture obtained by Atomic Force Microscopy. My investigations on this sample consisted basically in the same three approaches developed for the sample made of stripes of alternative width.

First, a 1 dimensional diffraction pattern was obtained in the scattering plane, as for the

³The pictures as well as the samples were provided by Unnar Arnalds of the Department of Materials Physics, Uppsala University.

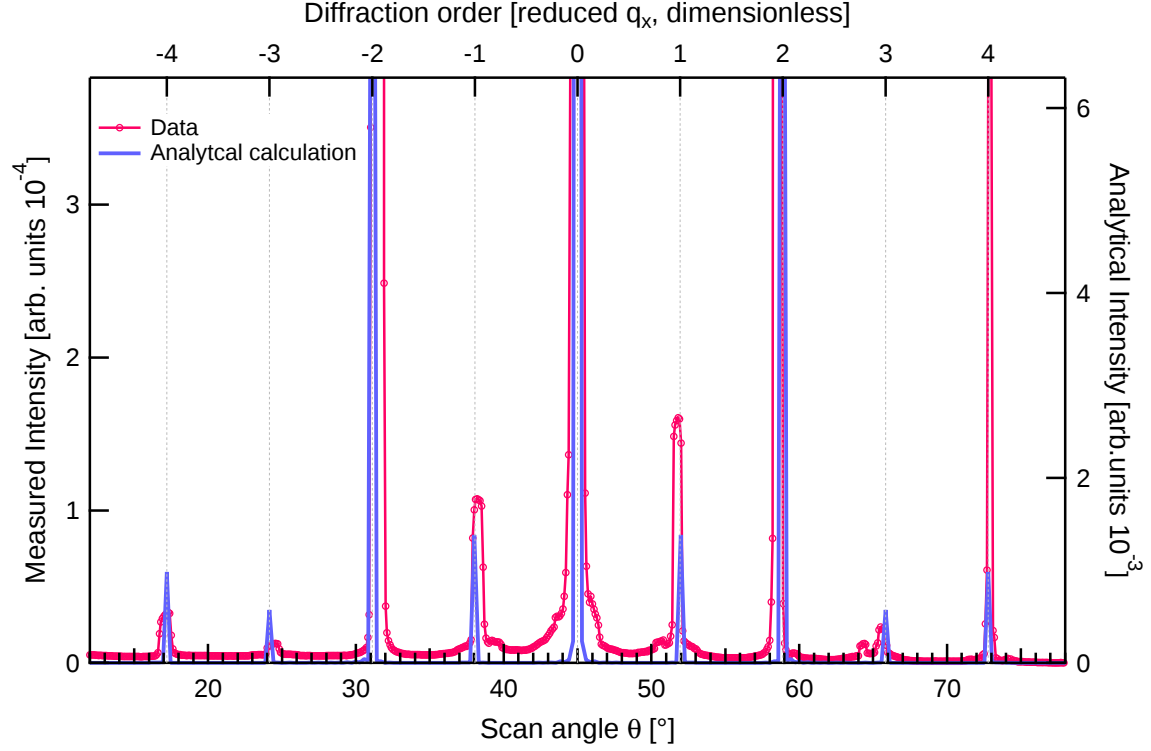


Figure 3.6: *Experimental and analytically obtained diffraction patterns of the sample made of alternate stripes (red and blue curve respectively). The patterns are shown in linear scale but cropped down to lower intensity values such that the high order diffraction rods can be seen and compared. The patterns are represented as functions of both the scan angle θ and the reduced coordinate q_x^R .*

first sample. It is shown in figure 3.9 by the reddish curve. Intensity is again represented as a function of the scanning angle θ .

Then, the sample is modelled in three dimensions, at first approximation as it as been described and as it can be interpreted to be when observed by AFM. The unit slab can be designed as a square with one eighth of sphere on each corners and half of an ellipsoid in a normal position in the centre. Figure 3.10 depicts the unit slab as it can be thought to be, at first approximation. It is computed by a 3 dimensional matrix which models the direct space as a mesh where rows are the x axis, columns the y axis and the matrix elements represents the hight of the surface, distributed continuously (in the limit of the discretization of the direct space) from 0 to 1. The comparison with the AFM picture is presented in figure 3.8b where the unit slab has been periodically repeated in the lateral directions of the surface.

As for the striped sample, a simulated diffraction pattern is computed and compared to the experiment. Only the structure factor is indeed computed, and its square modulus is presented in figure 3.9 along with the experimental data (intensity). The modulation of the intensity with respect to the scan angle θ , or equivalently the scattering vector q_x , shows strict analogy to the computed structure factor. This, given as an outlook, allows to conclude that the simulated patterned surface represents rather well the real patterned sample. , where the experimental diffraction pattern is schemed along with the simulated one.

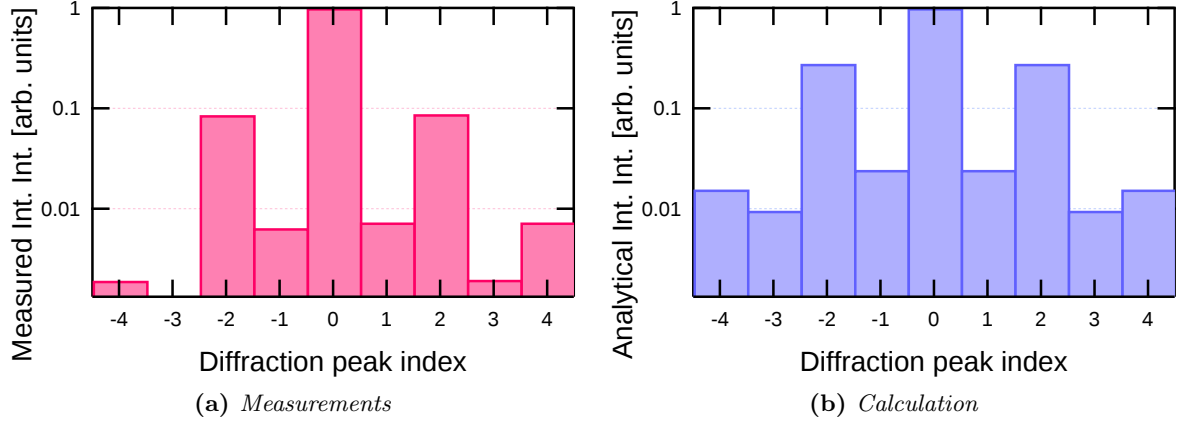


Figure 3.7: Comparison of the integrated intensities of both experimental and analytical diffraction pattern. Both are represented with the same logarithmic scale for comparison. Since the comparison of integrated intensity should allow to get rid of experimental considerations as far as comparing diffraction patterns is concerned, the quantitative differences should be assigned to a general mismatch between the model represented by the distribution functions and the actual sample. However, one notes a good qualitative agreement.

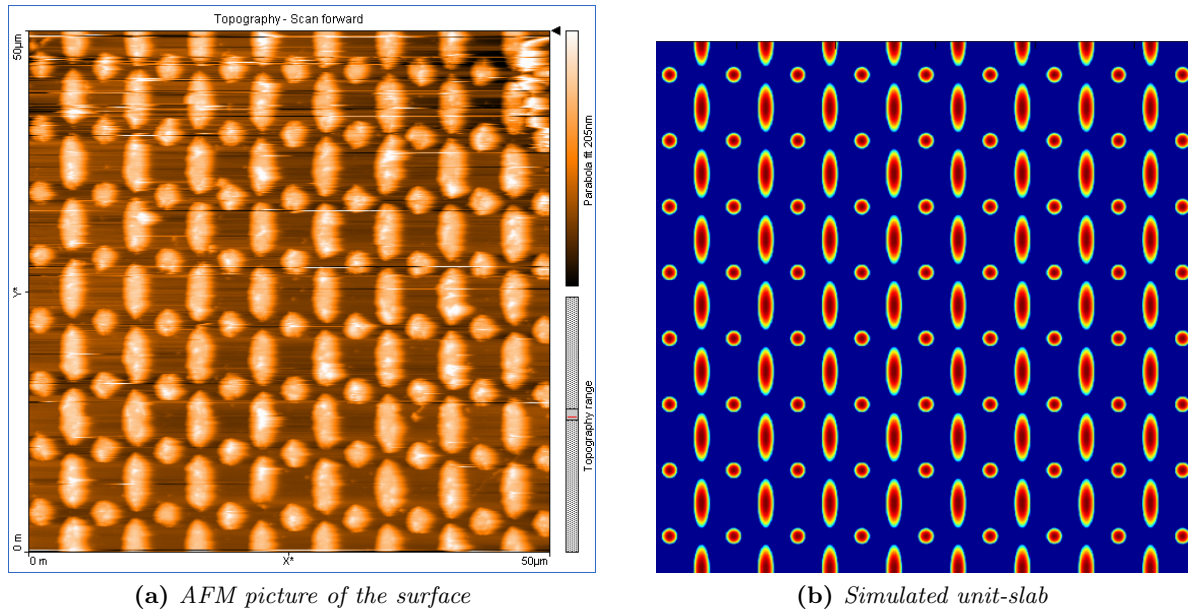


Figure 3.8: Atomic Force Microscopy pictures³ of the surface (left) of a patterned sample whose unit slab contains half of a spheroid shaped structure (four times a quarter of it) and half of an ellipsoid. The computed unit slab is depicted on the right.

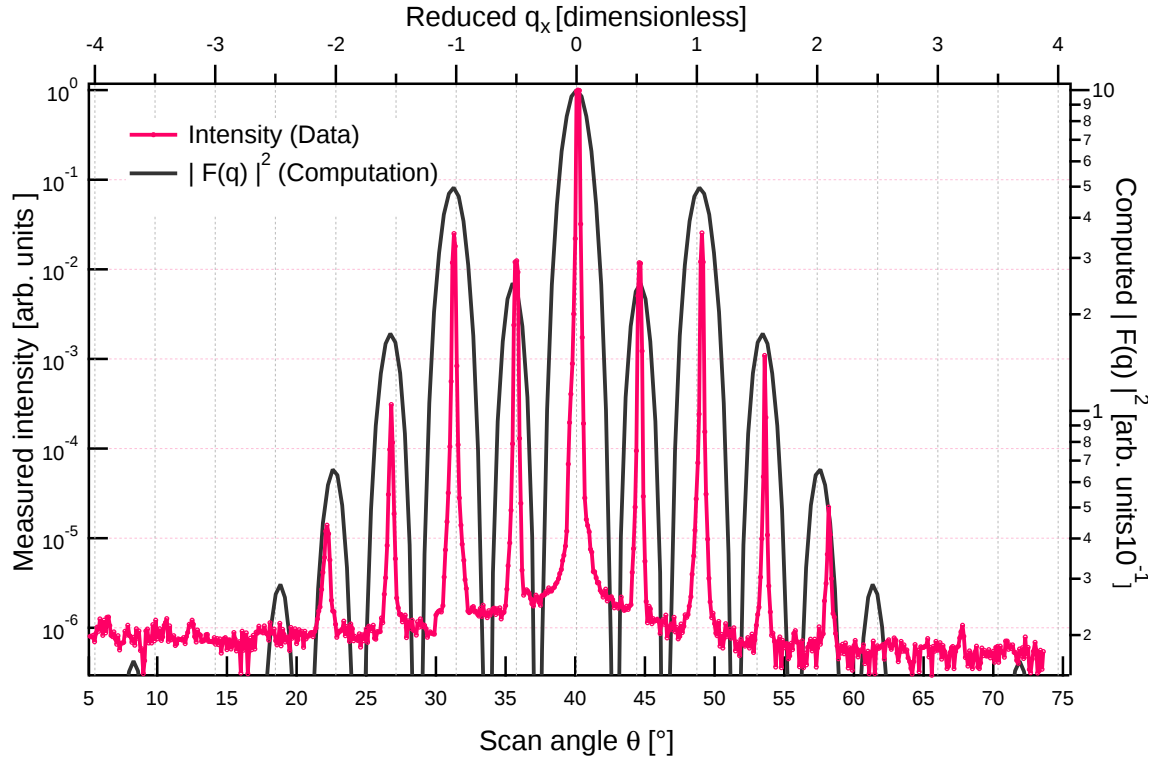


Figure 3.9: *Experimental diffraction pattern (intensity, red curve) compared to the computed structure factor (square modulus, black curve) in logarithmic scale as a function of both the scan angle θ and the reduced coordinate q_x^R . This gives a great insight on the possibility to model the patterned surface based on its diffraction pattern. The agreement of the computed square modulus of structure factor with respect to the actual diffraction pattern implies that the starting model is close to reality.*

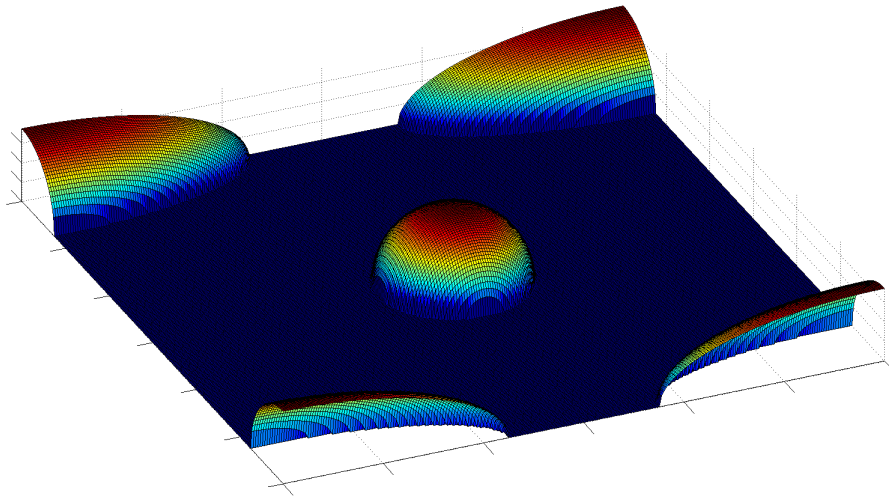


Figure 3.10: *Computational model of the unit slab in perspective view. The colours depicts the height of the individual surface structures.*

Conclusion

We have seen through this project course that optical diffraction made experimentally on patterned surface, coupled with an analytical approach based on simple theoretical models of the sample, which also served as the basis of simulations, is a powerful tool for structural characterization. The different approaches turned out to be very complementary and further studies, namely on the simulation part where geometric parameters can be varied at will, can give greater insight on how the actual studied sample surface is patterned. The simulation hence provides an additional tool that goes along the study of diffraction patterns which is based on known theoretical background now.

I would like to thank my advisor Matts Björck of the Department of Materials Physics of University of Uppsala, for his supervision all along this project and his support even afterwards. Also, I address my gratitude for Unnar Arnalds who designed the patterned samples that were investigated in this work. Additional thanks can truly be sent to Emil Melander who greatly helped me to use the set-up.

Appendix A

Mathematical derivations

A.1 Geometric sums

The geometric series is defined as a series with a constant ratio between successive terms. For any complex number r , the sum of the first N terms of a geometric series is:

$$s_N = \sum_{n=0}^{N-1} r^n = \frac{1 - r^N}{1 - r} \quad (\text{A.1})$$

This can be derived as follows:

$$\begin{aligned} s_N &= 1 + r + r^2 + \dots + r^{N-1} \\ \Leftrightarrow r \times s_N &= r + r^2 + r^3 + \dots + r^N \\ \Leftrightarrow s_N - r \times s_N &= s_N(1 - r) = 1 - r^N \\ \Leftrightarrow s_N &= \frac{1 - r^N}{1 - r} \end{aligned} \quad (\text{A.2})$$

The series converges for $N \rightarrow \infty$ if and only if $|r| \leq 1$, as shown below:

$$\begin{aligned} s_\infty &= \lim_{N \rightarrow \infty} s_N \\ &= \lim_{N \rightarrow \infty} \left[\frac{1 - r^{N+1}}{1 - r} \right] \\ &= \frac{1}{1 - r} \end{aligned} \quad (\text{A.3})$$

For the sums of phase factors that appear in the expressions of scattering amplitude 2.8 and 2.10, one has $r = e^{i\mathbf{q} \cdot \mathbf{r}}$ and the geometric series are of the following form:

$$s_N = \sum_{n=0}^{N-1} (e^{i\mathbf{q} \cdot \mathbf{r}})^n = \sum_{n=0}^{N-1} e^{i\mathbf{q} \cdot \mathbf{r} n} \quad (\text{A.4})$$

which can be evaluated, thanks to the above expression A.3, as:

$$\begin{aligned}
s_N &= \sum_{n=0}^{N-1} = \frac{1 - e^{i\mathbf{q} \cdot \mathbf{r} N}}{1 - e^{i\mathbf{q} \cdot \mathbf{r}}} \\
&= \frac{e^{-i\frac{\mathbf{q} \cdot \mathbf{r} N}{2}} - e^{i\frac{\mathbf{q} \cdot \mathbf{r} N}{2}}}{e^{-i\frac{\mathbf{q} \cdot \mathbf{r}}{2}} - e^{i\frac{\mathbf{q} \cdot \mathbf{r}}{2}}} \frac{e^{i\frac{\mathbf{q} \cdot \mathbf{r} N}{2}}}{e^{i\frac{\mathbf{q} \cdot \mathbf{r}}{2}}} \\
&= \frac{\sin\left(\frac{1}{2}N\mathbf{q} \cdot \mathbf{r}\right)}{\sin\left(\frac{1}{2}\mathbf{q} \cdot \mathbf{r}\right)} e^{i\mathbf{q} \cdot \mathbf{r} \frac{(N-1)}{2}}
\end{aligned} \tag{A.5}$$

A.2 Structure factor and intensity

The structure factor is computed starting from the expression provided in equation 3.2, and using the expression of the form factors for the two types of stripes, given in equation 3.3.

$$\begin{aligned}
F(\mathbf{q}) &= \int_{-\frac{\omega_1}{2}}^{\frac{\omega_1}{2}} e^{iq_x x} dx \int_0^h e^{iq_z z} dz + \int_{-\frac{a}{2}}^{\frac{\omega_2 - a}{2}} e^{iq_x x} dx \int_0^h e^{iq_z z} dz + \int_{\frac{a - \omega_2}{2}}^{-\frac{a}{2}} e^{iq_x x} dx \int_0^h e^{iq_z z} dz \\
&= \frac{1}{iq_x} [e^{iq_x x}]_{-\frac{\omega_1}{2}}^{\frac{\omega_1}{2}} \frac{1}{iq_z} [e^{iq_z z}]_0^h + \frac{1}{iq_x} [e^{iq_x x}]_{\frac{a - \omega_2}{2}}^{\frac{\omega_2 - a}{2}} \frac{1}{iq_z} [e^{iq_z z}]_0^h \\
&= \frac{1}{iq_x} 2i \sin\left(q_x \frac{\omega_1}{2}\right) \frac{1}{iq_z} (e^{iq_z h} - 1) + \frac{1}{iq_x} \left[2i \sin\left(q_x \frac{\omega_2 - a}{2}\right) + 2i \sin\left(q_x \frac{a}{2}\right) \right] \frac{1}{iq_z} (e^{iq_z h} - 1) \\
&= \underbrace{\omega_1 \text{sinc}\left(q_x \frac{\omega_1}{2}\right) \frac{1}{iq_z} (e^{iq_z h} - 1)}_{f_1(\mathbf{q})} + \underbrace{\left[(\omega_2 - a) \text{sinc}\left(q_x \frac{\omega_2 - a}{2}\right) + a \text{sinc}\left(q_x \frac{a}{2}\right) \right] \frac{1}{iq_z} (e^{iq_z h} - 1)}_{f_2(\mathbf{q})}
\end{aligned} \tag{A.6}$$

Then, the intensity is approached with the square modulus of the structure factor and the calculation of this term is performed by the computer, literally with $|F(\mathbf{q})|^2 = |f_1(\mathbf{q}) + f_2(\mathbf{q})|^2$.

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